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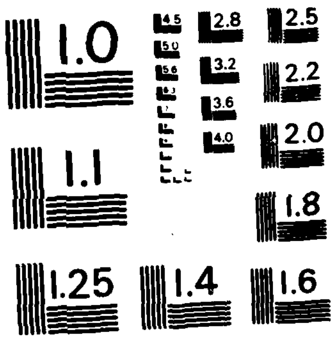

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Annual Technical Report

Contract AFOSR-83-0050

Solids Under Extreme Conditions

Dec. 83
Interim report
U. of Texas at Arlington

AD A139352

This report summarizes some of the significant results from the first year of research into properties of solids under extreme conditions: high pressures and low temperatures.

In the category of solids under high pressure the focus of this research has been a fundamental understanding of the ground and low lying excited states of explosive solids and energy transfer in these solids.

In the category of solids at low temperature, investigations included the following topics:

1. Coherent radiation from excited molecules at the surface of a metal.

2. Reformulation of the electron-phonon interaction in solids and calculation of the superconducting transition temperature.

3. Study of interaction of localized magnetic moments in metals and superconductors at low temperature.

4. Various transport properties of solids at low temperature.

5. Electron-phonon-magnon interactions.

6. Many body interactions in solids and at solid surfaces.

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Solid explosives, energetic materials, electron-phonon interactions, low temperature solid state effects, solids under high pressure

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

During the period Dec. 1, 1982 to Nov. 31, 1983 theoretical studies of energetic solids and solids at low temperature began. Major state-of-the-art quantum chemistry codes were implemented for the task of determining activation energies of explosive molecules and solids. Extensive studies of the R-NO_2 molecular species concluded the most previous work based upon a single determinant wavefunction is wrong: even the ground state of these molecules required a multi-configurational description. The generalized valence bond method has been employed successfully to describe a large number

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of molecules and offers hope of understanding some of the much more complex energetic molecules which are important because of their general use.

Studies of the electron phonon interaction have resulted in a new, efficient formulation of the problem. Theoretical prediction of the superconducting transition temperature base upon accurate description of the electron phonon interaction has been completed for a number of transition metals and their alloys. Studies of surface superconductivity and investigation of coherent radiation from molecules at solid surfaces have also begun.

I. Studies of Explosive Solids

This part of the progress report is concerned with studies of explosive molecules and the detonation process. A major part of the work that has been done in this regard is extensive computational studies of excited electronic states in R-NO_2 species. Characterization of these states is essential to a full understanding of the decomposition of these substances.

As a first step, we have made several major quantum chemistry codes[1] operational in this laboratory. During this process of conversion, it was noted that a major computational problem associated with studies of R-NO_2 species is that even the ground state of a simple molecule such as nitromethane requires a multi-determinant wavefunction for a reasonably accurate description.[2] In addition, many excited states can be expected to have multi-configurational character, as well as all states when the geometry is significantly displaced from equilibrium.

For reasons of computational efficiency, we decided to concentrate on the C_{2V} tautomer of nitrous acid, HNO_2. The molecule was chosen for two basic reasons: it is small enough to permit elaborate computational studies and it can be used as a model of alkyl-nitro compounds, which are typically explosive. In this molecule, the Hartree-Fock configuration is $|1a_1^2\ 1b_2^2\ 2a_1^2\ 3a_1^2\ 2b_2^2\ 4a_1^2\ 5a_1^2\ 3b_2^2\ 1b_1^2\ 4b_2^2\ 6a_1^2\ 1a_2^2|$, however, the configuration $|1a_1^2\ .\ .\ .\ .\ 6a_1^2\ 2b_1^2|$ mixes in strongly (the coefficient of this second configuration can be as high as 0.4 for a polarized double-zeta basis set). The conclusion that was drawn is that such a large contribution from a second configuration requires sophisticated correlation treatments such as MCSCF or GVB and that all nitro compounds require such treatments, in contridiction to several published works. We also found that minimal basis sets are simply

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inadequate for any reasonable description of the ground and excited electronic states of nitro compounds. Specifically, we calculated the energy separation between the ground and lowest excited triplet state as a function of basis set (3G, 3-21G and 3-21G + d's on N and O) and wavefunction type (single determinant SCF, SCF + two determinant CI, or two determinant MCSCF). The lowest triplet state is of $^3B_2$ symmetry and is properly described by a single determinant. Our results are given below:

	Correlation Correction (eV)			Singlet-Triplet Separation (eV)		
	3G	3-21G	3-21G*	3G	3-21G	3-21G*
SCF	-	-	-	-0.80	+0.69	+1.46
SCF +2 det CI	-2.05	-1.18	-0.91	+1.25	+1.86	+2.37
2 det MCSCF	-2.47	-1.64	-1.31	+1.66	+2.32	+2.77

The conclusions that we draw from this is that probably for all explosive systems: (1) Correlation corrections are very important. (2) At the SCF level, a minimum basis set incorrectly predicts the triplet to be the ground state. (3) SCF-CI and MCSCF treatments (with minimal basis set) underestimate the singlet-triplet separation and overestimate the correlation correction. (4) To peform accurate calculation, one must use a basis set of at least polarized double zeta quality and a correlation treatment such as MCSCF, followed by extensive CI treatments of both the ground and excited states.

At this point in our work, we were confronted with the impracticality of doing MCSCF + CI calculations on realistic molecules with the available computing facilities at UTA, because of the excessively high core and cpu requirements. We were, however, able to come up with a practical alternative--the main point is simply that realistic calculations on realistic molecules

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can be done within the Generalized Valence Bond (GVB) framework. This approach replaces one or more doubly occupied molecular orbitals with a valance bond like function, $\phi_\ell(1)\ \phi_r(2) + \phi_\ell(2)\ \phi_r(1)$, where the ϕ's are variationally determined. Please note that if there is only one GVB pair (fairly accurate for nitro compounds), GVB calculation is rigorously equivalent to a two determinant MCSCF. Extensive CI's can be done using the GVB wavefunction as the reference. Finally (in fact, one of the most practical considerations), the computational effort involved is significantly less. The one pair GVB calculation on HNO_2 with the 3-21G +d basis set requires only 30 minutes on an IBM 4341, compared to 171 minutes for the MCSCF. The two calculations yield exactly the same energy. During this process, after hours of computational effort, we were also able to partition the existing codes so that a GVB calculation can now be done with a 2M load module instead of a 7M load module for MCSCF calculations.

At this point in our work, we were satisfied with the computational aspects. We then concentrated on various R-NO_2 specifies, the first one is again HNO_2. We have presently, both 3-21G and 3-21G* results on HNO_2. However, for reasons of brevity, we only report the 3-21G results.

	Total Energy (a.u.)	ΔE (a.u.) (SCF)	ΔE (a.u.) (CI)
RHF	-203.42691		
GVB	-203.48704		
GVB-CI	-203.59087	0.025	0.134
ROHF	-203.40160		
ROHF-CI	-203.45660		

Please note that a significant amount of correlation is recovered using the CI treatment on both GVB and ROHF wavefunctions. These CI's are first-order CI's, it is our belief that a full CI is probably not necessary to accurately estimate (within 95%) singlet- triplet separations.

The next system we considered is nitromethane, an extremely well-studied molecule, by experimentalists and theorists alike. However, most of these calculations ignore the multi-reference character of the ground state of CH_3NO_2, thereby raising serious doubts about the validity of these studies. Our results are presented below:

State	Basis Set	Wavefunction	Energy	ΔE
$^1A^1$	3G	RHF	-240.4140	-0.48
$^3A^1$	3G	RHF	-240.4315	-
$^1A^1$	3G	RHF-CI	-240.4813	+1.83
$^1A^1$	3G	MCSCF	-240.4971	+2.26
$^1A^1$	3-21G	RHF	-242.2547	+1.06
$^3A^1$	3-21G	RHF	-242.2157	-
$^1A^1$	3-21G	RHF-CI	-242.2895	+2.01
$^1A^1$	3-21G	MCSCF	-242.3082	+2.52
$^1A^1$	3-21G	GVB-CI	-242.3082	+2.99
$^3A^1$	3-21G	ROHF-CI	-242.2751	-

Obviously, the table above raises questions about the suitability of semi-empirical methods for the study of the electronic structure of organic nitro compounds. Similar results have been obtained for HNO_3, NH_2NO_2, FNO_2 at the 3-21G level. At the polarized double-zeta level, the CI calculations are currently in progress. We also have preliminary results on CH_3ONO,

$ClNO_2$ and most notably, nitrobenzene. It is anticipated that by the end of the second year, we will complete all the relevant calculations on these systems and look closely into molecular clusters, e.g. $(CH_3NO_2)_n$ and other prototype systems.

II. Solids at Low Temperatures

A large number of projects are being conducted under this general category. The unifying idea is the search for special low temperature effects which might have device applications. In some of the projects a better understanding of well known phenomena is sought, e.g. the superconducting transition temperature, but the possibility of surface superconductivity is also being studied. While there are several projects here, not as much progress has been made as with part I, above. Some of this work has already been published, and a discussion of two of the topics is given below. Abstracts of talks give additional information.

1. Molecules at Surfaces

We are continueing to investigate the properties of excited molecules outside the surface of a metal. The decay rate of an excited molecule is enhanced due to the interaction of the molecule with its image charge in the metal. In addition, two or more excited molecules outside a metal surface interact with each other, creating a cooperative effect which produces a coherent decay of the excited molecules into the metal. The most efficient nonradiative decay channel is through the excitation of plasmon modes.[3]

We are presently in the process of comparing the results obtained by considering the coupling of a decaying molecule to the surface plasmons with those obtained by including the bulk plasmons modes. There are two reasons for including the bulk plasmons. First, as Babiker[4] has shown, if the quasistatic approximation is not invoked in the derivation of the interaction between a charge and a metal surface, the resulting retardation effects require that the image potential have a contribution from both the surface and the bulk plasmons. Second, in order to go beyond the usual

representation of the electron gas in a metal by a dispersionless dielectric function, spatial dispersion must be included. This is equivalent to the dependence of the dielectric function on the wave vector k. Barton[5] has argued that the usual decoupling of the bulk from the vacuum outside the surface does not apply when spatial dispersion is included.

Our present calculations are concerned with the interaction between an external Hydrogen atom and a metal represented by the jellium model. For the dielectric function representing the metal, we chose the hydrodynamical dielectric function, which Fuchs and Kliewer[6] have shown to be a good approximation to the more complicated Lindhard dielectric function. This dielectric function includes spatial dispersion. The contribution to the decay rate of the excited atom of the bulk plasmons is being calculated, and its importance will be evaluated. The next step will be to calculate the decay rate when two atoms are allowed to interact outside the metal.

The propagation of the resulting pulse of energy generated when a monolayer of excited molecules decays into the metal is being investigated. Morawitz[3] has predicted that this pulse will propagate with no energy loss; i.e., the pulse has the properties of a soliton. We plan to apply the coherent-state representation of a soliton, recently used by Isii[7] in another problem, to this analysis. We have applied this model to the calculation of the properties of positrons in metals.[8]

2. Electron-Phonon Interactions

During the period covered in this report, we have studied the problem of describing the electron-phonon (el-ph) interaction in solids using a modified tight binding scheme, and also developed artificial intelligence (AI) capabilities to help us investigate the problem. As a result of this

study we have established a new formalism for studying the el-ph interaction in transition metals. The AI programs have proved some of the necessary "theorems" and have derived the algebraic expressions for the el-ph interaction.[9] Using these programs one can and we are in the process of expanding our formalism to describe heavier elements by including f and g electrons. Also, contrary to the earlier assertion,[10] we have shown that one can successfully use the orthogonal tight-binding band structure and th. scaling laws for the Slater-Koster parameters developed by Harrisor :n describing the el-ph interaction. We think the conclusion of Varm t al., that it is necessary to use a non-orthogonal tight-binding band structure is unwarranted in light of errors in their work.

To us the long term significance of our study here, besides introducing AI to the study of el-ph interaction, is that the orthogonal Slater-Koster parameters are adequate to describe the el-ph interaction and since unlike the non-orthogonal ones they are fairly transferable from system to system[11] one can study the el-ph interaction in a variety of crystalline and non-crystalline solids and solid in different phases. This is particulary important for the study of the non-crystalline solids, since they are not amiable to the alternative Bloch formalism of the problem.

Also during this period we have used AI methods to improve the numerical stability of the norm-conserving pseudopotential.[12] In near future using the formalism we will persue the study of superconductivity of various solids and particularly surfaces, develop AI programs for study of el-ph in complex lattice structures, investigate the possibility of an unified theory describing magnetism, electron-phonon interactions and superconductivity.

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Cumulative Chronological List of Publications

The following papers have been published, submitted or are currently being written. All research is complete.

1. "Work functions and effective masses of positrons in metals," G. Fletcher, J. L. Fry and P. C. Pattnaik, Phys. Rev. B27, 3987 (1983).

2. "Improved numerical stability for norm-conserving ion-core pseudopotentials, P. C. Pattnaik, G. Fletcher and J. L. Fry, Phys. Rev. B28, 3564 (1983).

3. "Pressure dependence of the Fermi surface of paramagnetic chromium," P. C. Pattnaik, P. H. Dickinson and J. L. Fry, Phys. Rev. B28, 5281 (1983).

4. "The electronic structure of nitromethane: a multiconfiguration self-consistent field study," D. S. Marynick, A. K. Ray, J. L. Fry and D. A. Kleier, J. Mol. Structure (accepted for publication).

5. "Electronic structures of nitrons acid and nitromethane: generalized valance bond and configuration interaction studies," A. K. Ray, D. S. Marynick and J. L. Fry, submitted to J. Phys. Chem.

6. "Electronic structures of nitric acid and nitramine: generalized valence bond and configuration interaction studies," A. K. Ray, D. S. Marynick and J. L. Fry, submitted to J. Phys. Chem.

7. "Complete CI study of the vertical excitation spectrum of HNO," A. K. Ray and J. L. Fry, to be submitted to J. Phys. B.

8. "The use of algebraic programming in physics," P. C. Pattnaik and Glen Fletcher, submitted to American Journal of Physics.

9. "Electron-phonon interactions in transition metals," P. C. Pattnaik, M. E. Schabes and J. L. Fry,to be submitted to The Physical Review.

10. "A Study of the linear approximation in the Analytic Tetrahedron Method," P. C. Pattnaik and J. L. Fry (accepted for publication in Solid State Communications).

11. "K-dependence of matrix element in the Analytic Tetrahedron Method: Application to the orbital susceptibility of the uniform electron gas," J. L. Fry and P. C. Pattnaik (to be submitted to The Physical Review).

12. "Artificial intelligence for proving theorems in the study of electron-phonon interaction," P. C. Pattnaik, J. L. Fry and M. E. Schabes.

13. "Positron work functions in metallic alloys," P. C. Pattnaik and G. Fletcher (to be submitted to The Physical Review).

Interactions

1. "Quantum Size Effects in the Dielectric Function of Small Metallic Particles," (A. K. Ray and C. W. Myles), Bull. Am. Phys. Soc. 28, 284 (1983).

2. "Electron Transport in Layered Media by Invariant Imbedding," (A. K. Ray, P. Nelson and J. C. Garth), Bull. Am. Phys. Soc. 28, 428 (1983).

3. "The Electronic Structure of Nitromethane-A Generalized Valence-Bond Study," (A. K. Ray, D. S. Marynick and J. L. Fry), Bull. Am. Phys. Soc. 29, 62 (1984).

4. "Electronic Structure Calculations of Nitrous Acid," (D. S. Marynick, A. K. Ray, J. L. Fry and C. J. Liles), Bull. Am. Phys. Soc. (1984).

5. "Electronic Structures of Nitric Acid and Nitramine," (A. K. Ray, D. S. Marynick and J. L. Fry), Bull. Am. Phys. Soc. (1984).

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7. "The Electronic Structure of Nitromethane-A Configuration Interaction Study," (A. K. Ray, D. S. Marynick and J. L. Fry), Bull. Am. Phys. Soc. (1984).

8. "Pressure Dependence of the Fermi Surface of Paramagnetic Chromium," P. C. Pattnaik, P. H. Dickinson and J. L. Fry, Bull. Am. Phys. Soc. 28, 249 (1983).

9. "Spin-Lattice Relaxation of Conduction Electrons in Dilute Magnetic Alloys at Low Temperatures," J. L. Fry and N. G. Fazleyev, Bull. Am. Phys. Soc. 28, 256 (1983).

10. "Dynamics of Localized Magnetic Moments in Metals at Low Temperatures," N. G. Fazleyev and J. L. Fry, Bull. Am. Phys. Soc. 28, 325 (1983).

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12. "Electron Spin Resonance in Superconductors with Paramagnetic Impurities," N. G. Fazileyev, Bull. Am. Phys. Soc. 28, 505 (1983).

13. "Magnetic Resonance Lineshapes of Paramagnetic Ion Nuclei in Metals," N. G. Fazleyev, Bull. Am. Phys. Soc. 28, 349 (1983).

14. "Electron Transport in Layered Media by Invariant Imbedding," A. K. Ray, P. Nelson and J. C. Garth, Bull. Am. Phys. Soc. 28, 428 (1983).

15. "Quantum Size Effects in the Dielectric Function of Small Metalic Particles," A. K. Ray and C. W. Myles, Bull. Am. Phys. Soc. 28, 284 (1983).

16. "Fermi Surfaces of Ferromagnetic Iron and Nickel," J. L. Fry, S. S. Fry and C. F. Machala, Bull. Am. Phys. Soc. 28 (1983).

17. "Workshop on Many-body Phenomena at Surfaces," Institute for Theoretical Physics, University of California at Santa Barbara. Attended by Glenn Fletcher, August, 1983.

18. "A Study of the Linear Interpolation Used in the Analytic Tetrahedron Method," Bull. Am. Phys. 28, 196 (1983).

19. "Multipolar Interaction in Metals," N. G. Fazliyev and J. L. Fry, Bull. Am. Phys. Soc. 28 (1983).

20. "Electron Paramagnetic Resonance in Dilute Alloys at Low Temperatures," N. G. Fazleyev, Bull. Am. Phys. Soc. 28 201 (1983).

21. "Relaxation of Paramagnetic Ion-Nuclei in Type II Superconductors," N. G. Fazleyev, Bull. Am. Phys. Soc. 28, 204 (1983).

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23. "The Electronic Structure of Nitromethane: A Generalized Valence-Bond Study," A. K. Ray, D. S. Marynick and J. L. Fry, Bull. Am, Phys. Soc. 29, 62 (1984).

24. "Application of Artificial Intelligence to Calculation of the Electron-Phonon Scattering Matrix," M. E. Schabes, J. L. Fry and P. C. Pattnaik, Bull. Am. Phys. Soc. 29, 76 (1984).

25. "Surfaces Electron States on Cr(100)," G. Fletcher and P. C. Pattnaik, Bull. Am. Phys. Soc. 29, 62 (1984).

26. Workshop on Energetic Material Initiation Fundamentals, August 15-17, 1983 at Chestertown, Maryland (Department of Defense) was attended by J. L. Fry, A. K. Ray and D. S. Marynick.

27. Trip to Louisiana State University to consult with Professor J. Callaway on electron-phonon-magnon interactions and set up to use LSU Computers remotely. (Nov. 28-30, 1983) J. L. Fry and M. E. Schabes.

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